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Large Scale Tritium Recovery from Obsolete Illumination Devices at LLNL

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Since 2001, LLNL has supported a program to recover and recycle tritium from exit signs, telephone dials, gun sights and other military and commercial tritium-powered illumination devices. In addition to permitting tritium reuse, this effort also provides an environmentally safe disposal option. Recently, the startup of the Tritium Grinder System (TGS) in the LLNL Tritium Facility has added significantly to the program's capability and capacity. Actual results, including the collected gas chemical composition and unit processing rate are presented along with a summary of the design features, operating procedures and safety controls.

I. INTRODUCTION

Tritium recovery and recycle efforts at LLNL's Tritium Facility began in conjunction with its support of Department of Energy tritium consolidation and D&D initiatives in the 1990's, principally those at LLNL, Mound, KMS Fusion and Sandia California's Tritium Research Laboratory. The resulting expertise and reputation led to a 2001 US Army Joint Munitions Command request for assistance with its growing inventory of obsolete tritium powered illumination devices.

The years following the initial Work-for-Others (WFO) contract with the Army have seen a steady increase in both the number of requests for similar tritium recovery services and, importantly, the sheer number of devices to be processed. The recently completed Tritium Grinder System (TGS) gives LLNL the capacity to accommodate this increased demand.

II. TGS DESIGN AND PROCESS DESCRIPTION

The TGS was built to efficiently process tritium bearing illumination devices on a large scale. The TGS liberates gaseous tritium from these various devices by mechanically grinding (or shredding) them under vacuum in a modified industrial shredder built by SSI Shredding Systems. Liberated tritium is collected in a storage tank and periodically removed by trapping on a commercially available SAES getter.

II. A. Loading, Grinding and Tritium Collection

Figures 1 and 2 show the major components of the grinding and collection portion of the TGS. 'Baskets' of illumination devices to be processed are introduced through the Access Door on the far left side of the illustration and loaded on the conveyer belt (Fig. 3). When five baskets are positioned the access door is closed and the system pumped down. Argon purging is employed to minimize residual air and water vapor. After a suitable vacuum is attained all five baskets are shredded sequentially with debris accumulating in the Discharge Hopper. Strategically positioned cameras allow operators to follow basket progress up the conveyor and into the grinding chamber. Evolved gas is vacuum pumped to the Collection Tank. A series of purges with argon is then employed to ensure more complete tritium transfer.

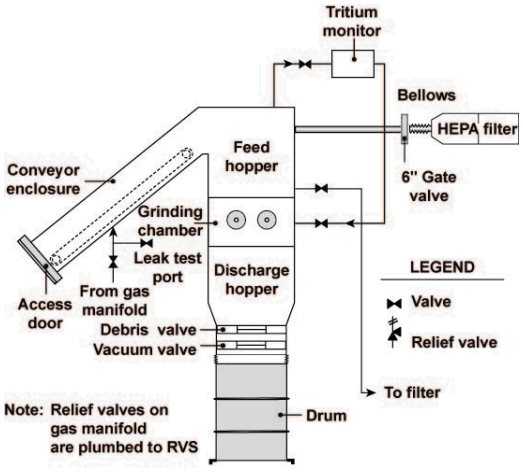

Fig. 1. TGS major components.

Fig. 2. TGS front view.

Fig. 3. Tritium handler loading phone dial baskets.

Following the final argon purge and pump out the TGS vacuum enclosure is let up to air, then vented to facility exhaust to permit tritium sampling, disposal of debris and conveyor reloading. A dedicated tritium monitor is consulted prior to venting to verify that not more than one curie will be released to building ventilation when opening the enclosure. A HEPA filter protects building ventilation from possible particulate formed during the shredding process and the building ventilation in turn provides an airflow protecting workers from residual contamination.

Solid debris generated by the recovery process is safely collected in a lined 55-gallon drum using a "bagout" technique similar to that developed for actinide gloveboxes. The bagout technique allows waste drum liners to be sealed and replaced without external exposure. Periodically a video camera and small vacuum cleaner nozzle are inserted through a small slit to inspect and clean the vital vacuum valve seal. Following closure the filled low level waste drum is set aside for later disposition and replaced with a fresh drum.

II.B. Tritium Purification, Storage and Disposition

Figure 4 schematically depicts the Tritium Recovery Loop portion of the TGS. The central feature is a high capacity SAES tritium getter (CapaciTorr-B, MK5 Type) The Recovery Loop is used to remove tritium (and other active gases) when Collection Tank pressure reaches 700 torr or 600 Ci, whichever is reached first.. Currently the relatively pure argon gas remaining in the collection tank after gettering is exhausted to facility ventilation, but the possibility of recycle is under consideration.

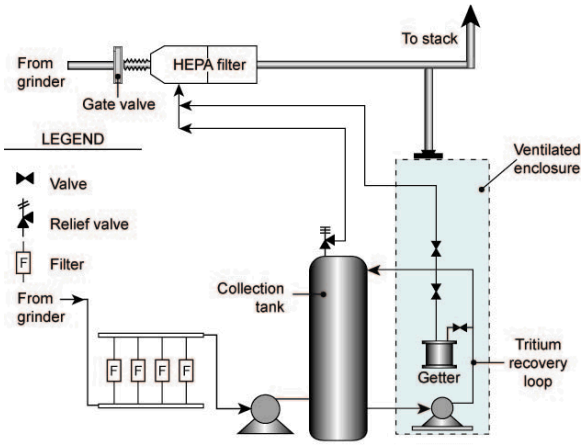

Fig. 4. TGS tritium recovery loop.

III. OPERATIONAL HISTORY

TGS operations began with a "Trial Period" designed primarily to gather information on the nature and type of radiological hazard the recovery process might pose. In Phase 1 very conservative assumptions were made about the nature and characteristics of tritium contamination that might be generated by the grinding process. In particular the possibility that residual fine particulates incorporating or otherwise contaminated with tritium, "Special Tritium Compounds" (Ref. 1), could become a worker hazard was taken into account by treating the area as an "airborne contamination area" and specifying a very high degree of worker protection. Worker protection measures included use of hooded Powered Air Personal Respirators (PAPR, 3M Model GVP-100) with tape sealed gloves and booties and extensive airborne particulate and surface swipe monitoring programs.

IV. TRIAL PERIOD RESULTS

Phase 1 Trial Period operations began on August 11, 2010 and concluded September 15, 2010 after ten grinding sessions that recovered tritium from 17,250 Gaseous Tritium Luminous Devices (GTLDs). GTLDs were used in British Telecom rotary dial telephones from 1964 to 1981 and consist of a tritium filled, horseshoe shaped glass tube glued onto an 84 mm diameter ABS plastic backing plate.

IV.A ES&H

IV.A.1. Tritiated Particulates

Breathing zone air samplers and continuous passive air samplers ("goose neck") monitors confirmed that airborne particulates were either extremely low in concentration or not present and thus not a problem for grinder operations with phone dials. The use of breathing zone samplers is thus discontinued for subsequent operations; passive air sampling will continue at least through the Phase 2 Trial Period. The highest work area posting is downgraded from "Airborne Contamination Area" to "Contamination Area"

IV.A.2. Spreadable Contamination

Consistent with past experience with manual processing of US Army illumination devices, moderate levels of surface contamination (2000 – 5000 dpm/100 cm^2) were found in several working area locations. Light decontamination (mopping) between grinding sessions has proven effective in keeping this spreadable contamination under control.

IV.A.3. Personnel Protective Equipment (PPE)

For Phase 2 use of a full face HEPA respirator is required only when the conveyor access door is open or while performing bagout related activities.

IV.B Performance Metrics

IV.B.1. Collected Gas Analysis

The Collection Tank pressure was 320 torr at the end of Phase 1. A mass spectrometric analysis of this collected gas is presented in Table I. The vast majority (99%) is of course the purge gas Ar. Air constituents O_2, N_2 and CO_2 are next, totaling 0.86%. That air is present is also unsurprising, though the O_2/N_2 fraction at 4.5, roughly the inverse of air, is interesting. This could, however, be due merely to differences in Ar sweep efficiency.

TABLE 1. Dial Gas Composition

Cmpd	Mole Fraction (%)
Ar	99
O2	0.67
N2	0.15
CO2	0.033
H2	0.066
3He	0.026
He	0.0085
HT	0.0035
HD	0.0032
D2	0.00078
T2	0.00045
DT	0.00036
Methanes	trace
Waters	trace
38Ar	trace
Total	**100**

Turning to the light isotopes, the preponderance of ^1H (protium) at 93% of total hydrogen is likely to primarily be the result of tritium exchange with environmental protium present as water vapor in the glass walls of the phone dial's tritium containment tubes.[2] Protium originating as H_2 within the glass probably also contributes.

The presence of deuterium can be explained as a known impurity in the original fill gas. Its total and relative (to ^3He and tritium mole fraction(s)) are broadly consistent with the known history of these dials. For example the 1.0 T_2/D_2 ratio is reasonable given that greater than 3 half lives of decay ('average' dial filled in 1972) have converted most of the original tritium fill to ^3He. After 38 years decay only ~ 12% tritium remains, suggesting an original deuterium fraction on this order, a reasonable result well within the significant uncertainties involved in this estimate.

The observed $^3He/T_2$ ratio of 10.5 is more difficult to interpret quantitatively due to hydrogen/helium permeation rate differences through the phone dial glass containment tubes and other factors. In the absence of permeation or other

differential loss mechanisms, however, a $^3He/T_2$ ratio of about 15 would be observed. This result suggests that permeation of helium is faster than tritium through the dial glass as would be expected.[3]

IV.B.2. Recovered Tritium

Phase 1 operations collected 58 Ci of recoverable tritium (as HT, DT or T_2) from an estimated 550 Ci total tritium associated with the 17,250 GTLDs processed. This 10.5% recovery rate compares well with the 12.5% expected from earlier measurements in the United Kingdom. The slight observed discrepancy may result from tritium labeled methanes and waters, known to be present in trace amounts but not yet accounted for quantitatively.

IV.B.3. Production Rate

From the results of Phase 1, in an average "production" day TGS operators can comfortably process three batches of 5 baskets each of low Ci illumination devices such as GTLDs. For higher Ci items such as exit signs (~ 5 Ci each) operations will likely be limited to 1/day by the need to remove tritium from the collection tank when it approaches 600 Ci.

ACKNOWLEDGMENTS

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